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Review of syngas production via biomass DFBGs

Kristina Göransson [a,*], Ulf Söderlind [a], Jie He [a], Wennan Zhang [b]

[a] Department of Natural Sciences, Engineering and Mathematics, Mid Sweden University, SE-871 88 Härnösand, Sweden
[b] Department of Natural Sciences, Engineering and Mathematics, Mid Sweden University, SE-851 70 Sundsvall, Sweden

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ABSTRACT

Production of high-quality syngas from biomass gasification in a dual fluidised bed gasifier (DFBG) has made a significant progress in R&D and Technology demonstration. An S&M scale bio-automotive fuel plant close to the feedstock resources is preferable as biomass feedstock is widely sparse and has relatively low density, low heating value and high moisture content. This requires a simple, reliable and cost-effective production of clean and good quality syngas. Indirect DFBGs, with steam as the gasification agent, produces a syngas of high content H_2 and CO with 12–20 MJ/m$_n^3$ heating value. A good quality syngas from DFBGs can be obtained by optimised design and operation of the gasifier, by the use of active catalytic bed materials including internal reforming of tars and methane, and finally by a downstream cleaning process. This article reviews the whole process from gasification to high quality syngas.

Contents

Abbreviations: AER, absorption enhanced reforming; BFB, bubbling fluidised bed; CFB, circulating fluidised bed; CLC, chemical loop combustion; DFB, dual fluidised bed; DFBG, dual fluidised bed gasifier; FB, fluidised bed; FT, Fischer and Tropsch; IGCC, integrated gasification combined cycle; LHV, lower heating value (MJ/m^3); MIUN, Mid Sweden University.

* Corresponding author. Tel.: +46 70 289 26 30/46 611 862 13; fax: +46 611 861 60.
E-mail addresses: kristina.goransson@miun.se (K. Göransson), ulf.soderlind@miun.se (U. Söderlind), jie.he@miun.se (J. He), wennan.zhang@miun.se (W. Zhang).

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1. Introduction

Bio-automotive fuels and chemicals can be produced from high-quality syngas (mainly H_2 and CO) which is obtained by gasification of biomass and wastes. This may be realised energy-efficiently and environmentally by integration with existing agricultural and forest industries, energy industry or chemical industry in a concept of biorefinery.

Gasification is an old thermo-chemical conversion technology with a change of the chemical structure of the biomass at 500–900 °C in the presence of a gasifying agent (for instance air, oxygen, steam, CO_2, or mixtures of these components).

The application of this combustible gas from coal, petroleum coke or biomass was commercial as early as 1812 [1], and the first attempt to use the gas for the internal combustion engine was carried out in 1881 [2]. Gasifers were widespread and used for industrial applications to power and heat. In Europe there were compact gasifier systems for automobile application developed between 1920 and 1940 due to the imminent Second World War and hence the unreliable petroleum supply. After the war, when the liquid fuels became available and inexpensive again, most of the gasifiers were decommissioned [1].

In the present situation however, there is a renewed interest in biomass gasification similar to the energy crisis of the 1970s, since a decreased use of fossil-based fuels is required. The world oil supplies are being depleted, and global warming is perceived as a threat to our environment. A number of gasifiers are under development and have the potential to produce a syngas suitable for bio-automotive fuel synthesis. Maybe, the dwindling oil supplies and global warming induce an energy revolution and hence initiate a bio century.

Gasifiers could be classified on the basis of gasification agent (air-blown, oxygen-blown or steam-blown), pressure (atmospheric or pressurised), temperature (slagging or non-slagging), fluid dynamics (updraft, downdraft, fluidised-bed or entrained flow) and in terms of heat supply (indirectly or directly heated) [3].

Direct gasification (or *autothermal* gasification) occurs when the oxidant gasification agent partially oxidises the feedstock and provides the heat for the process. Low temperature direct gasification (below 900 °C) can be carried out in a fixed bed, a fluidised bed (FB) or a circulating fluidised bed (CFB), and high temperatures direct gasification (over 1300 °C) in an entrained flow gasifier. Direct gasification with air produces a fuel gas of heating value 4–7 MJ/m_n^3— not suitable for synthesis of bio-automotive fuels. Pure oxygen gasification generates a fuel heating value of 10–12 MJ/m_n^3, but an oxygen plant is needed, which can be economic only for large-scale bio-automotive fuel production plants [4].

Entrained flow gasification technology has been well commercialised in large-scale integrated gasification combined cycle (IGCC) coal power plants. In most cases, the gasifiers are operated under pressure (typically 20–50 bar) with pure oxygen and with capacities in the order of several hundreds of MW_{th}. The syngas contain little or no methane and other hydrocarbons [3]. The technology is being demonstrated for black liquor gasification. The key issue in black liquor entrained flow gasification is the melting inorganic chemicals, while the key issue in biomass gasification is tar/CH_4 treatment. Biomass gasification and black liquor gasification are two different fields in the sense of scientific research and development. The world production of black liquor is about 670 TWh/year, fairly little potential in comparison to biomass [3].

For biomass however, an small or medium scale bio-automotive fuel plant close to the feedstock resources is preferable as this kind of feedstock is widely sparse and has relatively low density, low heating value and high moisture content [5]. This requires a simple, reliable and cost-effective production of clean and good quality syngas.

Indirect gasification (or *allothermal* gasification) produces a syngas of 12–20 MJ/m_n^3 heating value. The process does not occur with air as oxidising agent, it employ an external energy source for the necessary gasification heat. Dual fluidised-bed gasifiers (DFBGs), with steam as the gasification agent, turns out to be a promising biomass gasification technology. Steam is a commonly used indirect gasification agent. It is easily produced and increases the hydrogen content of the syngas. The gasifier consists of two separate reactors: a steam gasifier that converts feedstock into syngas, and a combustor with air that oxidises the residual char and hence provide the necessary heat to gasify the feedstock. The indirect DFBG is a combination of two fluidised beds, typically a BFB and a CFB.

Comparing to auto-thermal pressurised gasifiers, the advantages of a DFBGs are [3]:

- no oxygen demand to obtain nitrogen-free syngas;
- low investment costs;
- no or simple pre-treatment of biomass;
- easy feeding of biomass;
- suitable for biomass-based S&M scale bio-automotive fuel plant;
- low temperature operation;
- technology has been developed and demonstrated for heat and electricity production.

There are also other kind of indirect gasifiers, for instance, the gas indirect gasifier that burns a fraction of the syngas in heat exchange tubes surrounded the gasifier to provide the gasification heat. This review paper will focus on the production of high-quality syngas for automotive fuels synthesis from biomass gasification in a DFBG.

2. Dual fluidised bed gasifiers

DFBGs have been used for three purposes: (1) to supply heat for gasification (the common), (2) to supply oxygen (the Chemical Loop Combustion (CLC) process), and (3) to capture CO_2 (the Absorption Enhanced Reforming (AER) process). The CLC process employs a metal oxide as a bed material, and renders a redox cycle possible. The AER process uses limestone (CaO) as sorbent for in situ CO_2 capture during gasification [6]. The concepts are sketched in Fig. 1.

The second and third concepts are described in detail in the next section.

For the first concept, used most commonly, one bed is blown with steam to gasify biomass and to produce syngas at 700–900 °C; the other bed is blown with air to burn char to produce heat. The bed material as the heat carrier is circulated between these two beds to transfer heat from the combustion bed to the gasification bed, while separating the flue gases in the combustor from the syngas in the steam gasifier. DFBG produce two gases: a syngas with little or no nitrogen and a flue gas.

The bed material, together with the char left over from steam gasification, is circulated to the combustion bed. The heat produced in the combustor must be sufficient to manage the heat loss from the gasifier and the thermal energy requirements of the gasification reactions. The temperatures of the gasifier and the combustor are maintained by the char combustion and the bed material recirculation.

A lower gasifier temperature leads to higher char yield, and subsequently results in higher combustor temperature, hotter bed material and higher gasifier temperature, so that less char will be produced in the gasifier. Thus a balance of temperatures between the gasifier and the combustor can be reached—the system is auto-stabilising. The available DFBGs are summarised in Table 1.

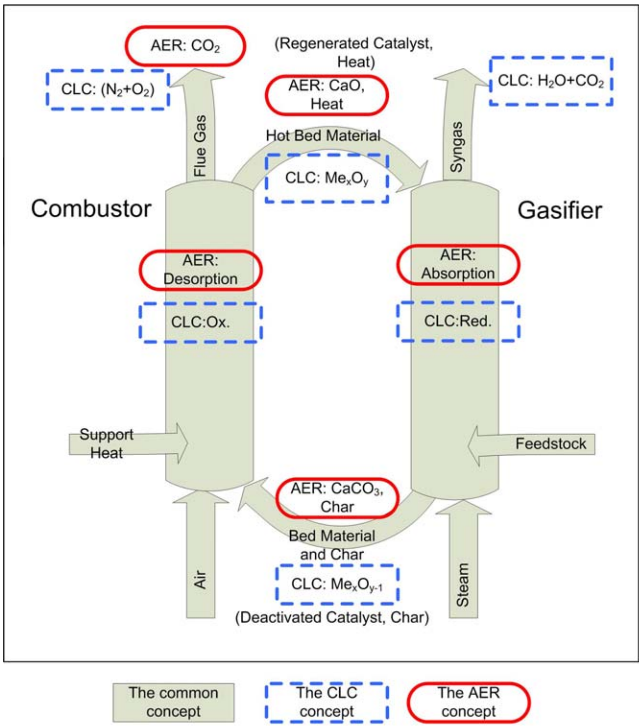


Fig. 1. Three concepts of the DFBG gasification.

The location where the biomass is fed influences its heating rate. It is most favourable to feed the biomass directly into the fluidised bed of the gasifier [22]. The syngas yield is significantly increased by in-bed feed of biomass compared to feeding above the bed in the gasifier [14].

The gas distributor plates in a fluidised bed require a correct design to prevent back flow of bed material through the nozzles [14]. Furthermore, the plate inclination is of significance. The produced char would go towards the upper zone of the bed in a fluidised-bed gasifier with a horizontal gas distribution plate. An inclined plate, however, results in a revolving circulation in the bed and hence a mixture of the solids. This benefits further gasification and also the transfer of the char towards the combustor. Unfortunately, the overall conversion of the H_2O will be low, since an inclined plate causes a low bed height of one side of the gasifier with a small residence time of the H_2O flow [22].

The temperature balance in the DFBG can be controlled by the bed material circulation, which is an important parameter to study, especially when a temperature difference between the gasifier and the combustor is required, such as in the case of the AER (adsorption enhanced reforming) process.

There are some technical options for DFBG technology due different combinations of the BFB and the CFB. So far, the most attractive design is supposed to have biomass gasification in the BFB and char combustion in the CFB from the viewpoint of particle circulation, fuel conversion and tar production [23]. Examples of this design are the Güssing gasifier e.g. [24–27] and the gasifier at Mid Sweden University (MIUN).

The MILENA gasification process (optimised for the production of substitute natural gas (SNG) from biomass) uses the riser for gasification and the BFB for combustion [9].

Gasifiers could be atmospheric or pressurised. At present, indirect DFBGs operate near atmospheric pressure. A pressurised gasifier results in a smaller volume of produced gas into the cleaning system. Furthermore, downstream synthesis processes often require pressurised gas. Thus, a compressor may not be required, leading to electricity saving. Positive effects of the pressure on the biomass gasification have been shown by several authors:

- the rate constant of steam-char gasification reaction increases with increasing steam pressure [28];
- the constant of the water gas shift reaction increases;
- the efficiency of gas phase reactions is enhanced [29];
- the reforming of tar and methane is enhanced.

However, under high pressure, biomass is difficult to be fed, and the capital and operation costs are raised significantly.

Advantages and disadvantages of pressurised gasification process in comparison to the atmospheric process are summarised in Table 2.

A pressurised (up to 10 bars) fluidised-bed gasifier was built at the Institute of Chemical Engineering at the Vienna University of Technology, to investigate the pressurised gasification process, which is their first step towards application of pressurised DFBGs. The pressurised CFB gasification has been demonstrated in Värnamo [30,31]. Solid fuel combustion in fluidised beds at elevated pressure is well established and common known. The BFB and CFB gasification technology is promising when the system is pressurised to facilitate syngas cleaning and downstream syntheses reactor [3].

3. Bed materials

The main function of the bed material in a DFBG is to supply the gasification zone with energy for the conversion of the biomass. It can make in situ gas conditioning in the same time. Reactive bed materials can be applied to improve agglomeration behaviour, to enhance tar cracking and to increase H_2 content. Moreover, the bed material can be used to perform catalytic activity, CO_2 capture, oxygen transportation, etc.

3.1. Catalytic effects

Catalytic reforming can be performed either with catalytic bed materials during gasification or after gasification using a separate, downstream catalytic reactor. Catalytic bed materials promote char gasification, water-gas-shift and steam reforming reactions, and reduce the tar yield. There is a great interest in in-bed additives use and the need for complex downstream cleaning methods can be simplified [32].

Examples of natural minerals are dolomite, limestone, olivine and iron ores, and examples of synthetic minerals are Ni-supported olivine, Fe-supported olivine, alkali metal-based material, and char [33]. Sulphur (H_2S and COS) can be retained in the bed (up to 90%) by sorbents such as limestone [34]. An inexpensive material for tar cracking can be char. But, as char itself gets converted during the process, an external continuous supply of char into the gasifier is required [32].

Although many in-bed catalysts showed good results concerning syngas quality, some of them are very expensive and some leave contaminated residues behind [22]. Additionally, there have been attrition problems and loss of catalytic activity. Ni-based catalysts are quickly deactivated due to carbon deposition and sulphur poisoning [32]. Good quality additive still remains to be found.

3.2. Internal reforming

The biggest challenge for fluidised-bed gasification is the reforming of tars and CH_4. Thermodynamic efficiency losses can be reduced using catalytic bed materials in the gasifier. Internal reforming retains the chemical energy of tars and methane in the

Table 1
Available biomass DFBGs.

Name/location/operation start	Capacity as fuel input (MW_{th})	Feedstock	Design (gasifier/combustor)	Bed material	Temperature (gasifier/combustor, °C)	Syngas composition (vol.%, dry)	Tar content (raw gas, g/m_n^3)	Cleaning process
Güssing FICFB/Austrian Energy, TU Vienna/ 2001 [7,8]	8	Biomass chips	BFB/CFB	olivine	900/1000	CO: 20–30 H_2: 35–45 CO_2: 15–25 CH_4: 8–12 N_2: 3–5	1.5–4.5	Fabric filter and biodiesel scrubber
Milena/ECN Netherlands/ 2004 [9]	0.03	Beech wood chips	CFB/BFB	Sand	850–900/925	Raw gas: CO: 39.3 H_2: 21.4 CO_2: 13.9 CH_4: 12.8 N_2:?	32	OLGA gas cleaning sector, catalytic reactors
Milena/ECN Netherlands/ 2008 [10]	0.8	Wood pellets	CFB/BFB	Sand	850/925	CO: 37–39 H_2: 18–20 CO_2: 11–13 CH_4: 14 N_2:?	40	See above
Chalmers/(GoBiGas) Sweden/ 2008 [11]	2	Wood pellets	BFB/CFB	Sand	812/?	CO: 33.1 H_2: 25.1 CO_2: 14.8 CH_4: 11.8 N_2: 9.3	7.8	?
MIUN/Mid Sweden University/2007	0.15	Wood pellets	BFB/CFB	Sand	800/950	CO: 35 H_2: 46 CO_2: 10 CH_4: 11 N_2: 4	10–65	no
SilvaGas/Vermont, USA/1998 [12]	~40 (design basis) ~90 (demonstrated capacity)	?	CFB/CFB	Sand	–	CO: 50 H_2:15 CO_2: 10 CH_4: 15 N_2:?	?	?
CAPE FICFB Gasifier/ University of Canterbury, New Zealand/2006 [13–15]	0.1	Wood chips/pellets	BFB/CFB	Sand	753/?	CO: 28.4 H_2: 21.7 CO_2: 17.4 CH_4: 11.6 N_2: 16.9	"Medium"	?
The JOULE–MFCF PlantENEA Trisaia, Italy/? [16]	0.5	Almond shells biomass	BFB/CFB	Catalytic bed	?	CO: 25.1 H_2: 33.1 CO_2: 19.3 CH_4: 10.4 N_2: 9.6	10.6	Adsorbing reactor and hot gas filter
Blue Tower/Herten, Germany/2001 [17,18]	15	Wastes	–	Ceramic Balls	600/950	CO: ~20 H_2: ~50 CO_2: ~20 CH_4: ~5 N_2:?	Low	?
?/Yokohama, Japan/? [19]	~0.02	Coffee grounds	BFB/CFB Two-stage DFBG	?	820/?	CO: 29.20 H_2: 31.23 CO_2: 17.28 CH_4: 13.68 N_2:?	~27	?

Table 1 (*Continued*)

Name/location/operation start	Capacity as fuel input (MW_{th})	Feedstock	Design (gasifier/combustor)	Bed material	Temperature (gasifier/combustor, °C)	Syngas composition (vol.%, dry)	Tar content (raw gas, g/m_n^3)	Cleaning process
?/National Institute of Advanced Industrial Science and Technology (AIST), Japan/? [20]	~0.0003	Cedar/White oak	BFB/BFB	Porous γ-alumina particles	800/~930	CO: ~26/29 H_2: ~40/34 CO_2: ~28/23 CH_4: ~9/9 N_2:?/?	?	?
?/ECCMB Dalian University of Technology, China/? [21]	~0.005	Pine sawdust	Downflow-Moving Bed/CFB	Calcined naturally occurring olivine	800/900	CO: 26–30 H_2: 40–44 CO_2: 16–23 CH_4: 9–13 N_2: ?	~2–3	?

syngas. An innovative concept of integrating a catalytic hot gas filter in the freeboard of a fluidised-bed steam gasifier has been tested by Rapagná et al. They used Ni catalyst filled into a hollow-cylindrical silicon carbide based filter [35]. The average variation of the content in the produced gas showed an increase of the hydrogen yield by 130% and a decrease of the methane and the tar content by 20% and 79% respectively. Furthermore, catalytic hot gas filter (monolith converters) can be placed at the gasifier outlet.

3.3. CLC

CLC was primarily proposed to increase thermal efficiency in power generation stations. It turned out to offer the inherent feature of isolating CO_2 with minimum need for costly separation processes, and in addition, formation of NO_x is reduced.

A CLC system is composed of two reactors: an air and a fuel reactor. The bed material is circulated between these two reactors (see Fig. 1). By reduction of the bed material in the fuel reactor (1) and subsequent oxidation in the air reactor (2) the combustion air and the fuel are never mixed. CLC can be used as partial combustion of the fuel in DFBG (cf. gasification with addition of pure oxygen) to increase the temperature and thereby reducing the tar and methane content.

$$(2n+m)Me_xO_y + C_nH_{2m} \rightarrow (2n+m)Me_xO_{y-1} + mH_2O + nCO_2 \quad (1)$$

$$Me_xO_{y-1} + 1/2O_2 \rightarrow Me_xO_y \quad (2)$$

Taking reactions (1) and (2) together, the fuel has been combusted but the resulting CO_2 has been separated from the N_2 in the air. Depending on the metal oxide reaction (1) whereas is often endothermic, reaction (2) is always exothermic [36].

3.4. AER

The aim of the AER (absorption enhanced reforming) process is to achieve high hydrogen content in the product gas.

The CO_2 adsorbent bed material used in the AER process has two purposes: firstly as a heat carrier and secondly as an in situ CO_2 capture. The CO_2 is transferred from the gasifier to the combustor (see Fig. 1) according to reaction (3)—a cyclic carbonation of CaO and calcination of $CaCO_3$.

$$CaO + CO_2 \Leftrightarrow CaCO_3 \quad (3)$$

The driving force for the carbonation and calcination is the difference between the partial pressure of CO_2 in the reactor and the equilibrium partial pressure of CO_2 [6]. Because CO_2 is removed inside the gasifier CO_2 and CO contents are deduced and a H_2 rich gas is obtained [37]. The use of CO_2-adsorbing bed materials within DFBGs render a product gas with hydrogen contents up to 75 vol.% possible [38]. The continuous CO_2 removal during the gasification enhances the reforming reactions and the WGS-shift reaction (4) towards H_2 production, since the change of CO_2 concentration drives the reaction to the right [6].

$$CO + H_2O \Leftrightarrow CO_2 + H_2 \quad (4)$$

The absorption process in the gasification zone is exothermal and generates heat for the gasification. Therefore a heat exchanger is necessary in order to control the temperature. Moreover, the AER process requires increased bed material residence times for the absorption/desorption of CO_2, which can be achieved by larger bed material particles. This in turn leads to increased steam volume flows to maintain the fluidised-bed regime [38].

Table 2
Advantages and disadvantages of pressurised gasification process in comparison to the atmospheric process.

Advantages	Disadvantages
A considerable increase of DFBG capacity, which is essential for synthetic fuel production	Less experience in syngas production
A smaller volume of syngas into the cleaning system	Difficult pressure balance between gasifier and synthesis
The syngas require less or no compression—i.e. saving cost for electricity	Significantly higher capital costs and operating costs
Energy efficiency advantages in combination with high temperature gas cleaning	The complex high temperature gas cleaning is under development
Reduction of the plant space	Fuel feeding problems
A lower surface of the gasifier gives a lower heat loss	Higher inert gas consumption during operation
Improved heat transfer in the bed	Difficulty in experimentation

Table 3
Indicative composition of syngas from an indirect gasifier [3].

Content	Vol.%, dry
H_2	40
CO	25
CO_2	21
CH_4	10
C_2H_4	2.5
N_2	1.5
H_2/CO	1.6
LHV (MJ/m^3)	14

Table 5
Specifications of maximal concentration of impurities in syngas for FT catalysis processes [41].

Impurity	Specification
$H_2S + COS + CS_2$	<1 ppm
NH_3 + HCN	<1 ppm
HCl + HBr + HF	<10 ppbV
Alkaline metals (Na + K)	<10 ppbV
Particulate	Essentially removed
Organic compounds (tars)	Below dew point
Hetero-organic compounds (Incl. S, N, O)	<1 ppmv

3.5. Increase of the ash melting point

From the viewpoint of low tar content in a high-quality syngas, the temperature in the bed should be as high as possible. However, the gasification temperature in a fluidised-bed cannot exceed the softening temperature of the ash. There is a risk that low-melting compounds or low-melting mixtures of particles are formed and deposit themselves on the bed particles. This can coat them together and form large agglomerates that may result in defluidisation of the bed.

These problems in fluidised-beds are usually associated with a high content of alkali metals in the fuel, especially in combination with a high content of sulphur, chlorine, silica and phosphorus since these elements form eutectics, which coat them with a sticky ash layer. Alkali compounds together with the silica from silica-sand bed can form low-melting surfaces on the particles, which promote agglomeration [39].

The addition of high-melting minerals (such as CaO or MgO), alkali-sorbents (clay minerals) and sulphur sorbents (such as limestone and dolomite) in the bed material, prevents the agglomeration tendencies in fluidised-beds. However, in some cases reported sulphur-sorbents decreased ash deformation temperatures and increased fouling [39].

4. Specification of typical syngas

4.1. The syngas property

The syngas property from a typical indirect gasifier is listed in Table 3. It contains H_2, CO, CO_2, CH_4, H_2O, trace amounts of higher hydrocarbons, possible inert gases present in the gasification agent and various contaminants. The content of H_2 may be further increased and the syngas can be improved using an in-bed CO_2 adsorbent and additives or catalysts. Some examples on contaminant present in the syngas are listed in Table 4.

There has been much experience gained from gas cleaning related to engine and turbine applications. However, product gas for synthesis normally has a much stricter specification of impurities than these applications. The published target levels which had to be met for all particulate and gaseous contaminants are extremely low, far below 1 ppm.

For instance, the catalysts in the Fischer–Tropsch synthesis (Fe) or the methanol synthesis (Cu) are sensitive to contaminants, mainly sulphur, chlorine and nitrogen compounds [40]. Catalysts for synthesis can be poisoned by NH_3, HCN, H_2S and COS. HCl causes corrosion, and alkaline metals and tars can be deposited on the catalysts, which results in the poisoning of catalysts and the contamination of the products. Moreover, particles like dust, soot and ash cause fouling of the reactor [3].

Maximal concentration of impurities in syngas for FT catalysis processes are specified in Table 5.

Table 6 summarises desirable syngas characteristics for different end products.

4.2. Effects of operation parameters on syngas composition

The composition of syngas is mainly dependent on the type of gasification agent used. A higher S/B (steam-to-biomass) ratio means a higher steam partial pressure that enhances the water-gas shift reaction to the right for more H_2 production at the expense of CO.

A higher gasification temperature will be in favour of the H_2 and CO yields since the endothermic gasification reactions are enhanced. Also from the viewpoint of low tar content, the

Table 4
Contaminant presence in the syngas.

Contaminant	Problems
Alkali metals	Can cause high-temperature metal corrosion and defluidisation of the bed. Alkali metals exist in vapour phase.
Fuel-bound nitrogen	Formes NO_x during combustion and causes potential emissions problems. Furthermore, the catalysts are sensitive to nitrogen compounds.
Particulates	Cause erosion of metallic components and environmental pollution. Originate from ash, char, bed material and condensing compounds.
Sulphur and chlorine	Could cause dangerous pollutants and acid corrosion of metals. The catalysts are sensitive to sulphur and chlorine compounds.
Tar	Clogs filters and valves and produces metallic corrosion. Tars exist in vapour phase in the syngas.

Table 6
Syngas composition required by various synthetic fuels.

Product	Pressure (bar)	Temperature (°C)	H_2/CO	Comment/catalyst
FT fuels	~20–40 [3] 25–40 [42]	200–240 [3]	0.6 for iron catalyst, 2.0 cobalt catalyst [43]	Production of high molecular mass linear waxes [3]
		300–350 [3] 250–350 [42]	0.6 for iron catalyst [43] 1.5–2 [42]	Production of gasoline and linear low molecular mass olefins [3] Fe/Cu/Mo [42]
Methanol	50–100 [3] 20–50 [44] 50–300 [42]	100–200 [43] 200–300 [3] 210–290 [44] 220–450 [42]	2.0 [43] 2.0 [42]	Copper–zinc-based catalysts [3] Zn/Cr, Cu/Zn [42]
DME	26 [3] 20–50 [44]	310 [3] 210–290 [44]	Converted from methanol [3]	Special alumina [3] catalysts
Ethanol	100 [3]	275 [3]	1.0 [3]	Supported rhodium catalysts [3]
Hydrogen	~28 [43]	100–200 [43]	High [43]	
Bio SNG	200–500 [42]	300–450 [42]	3.0 [42]	Ni/Mg [42]

temperature in the bed should be as high as possible. This is not easy in terms of biomass gasification with pure steam. Furthermore, high alkali content in the biomass limits the maximum allowable gasification temperature, since the alkali content influences the softening temperature of the ash generated in the gasification, with subsequent agglomeration problems [22]. Longer residence time of the gas in the bed would increase the tar cracking, but this requires a higher bed height if the superficial gas velocity in the bed should be unchanged.

Regardless of temperature in the bed of the gasifier, it is possible to reduce the tar yield and improve the product distribution from the gasifier by increasing S/B. However, this also means that the conversion of the H_2O in the gasifier gradually decreases; a greater proportion of steam flows through it unused.

The NH_3 content in the syngas is mainly determined by the content in N-containing species in the biomass [34].

5. Gas cleaning

Syngas can be cleaned to different degree depending on a balance of economic cost against technical specification for downstream synthesis. A cleaning process is generally required for a high-quality syngas for automotive fuels synthesis. The gas cleaning can be divided into conventional gas cleaning and dry hot gas cleaning. Tar treatment is described in next section. An overview of the cleaning processes can be seen in Fig. 2.

5.1. Conventional gas cleaning

Conventional gas cleaning, i.e. cold wet gas cleaning, is a proven technology but is thermally inefficient and produces waste water sludge, in other words, there is a scope for further research to develop dry, cold cleaning [51].

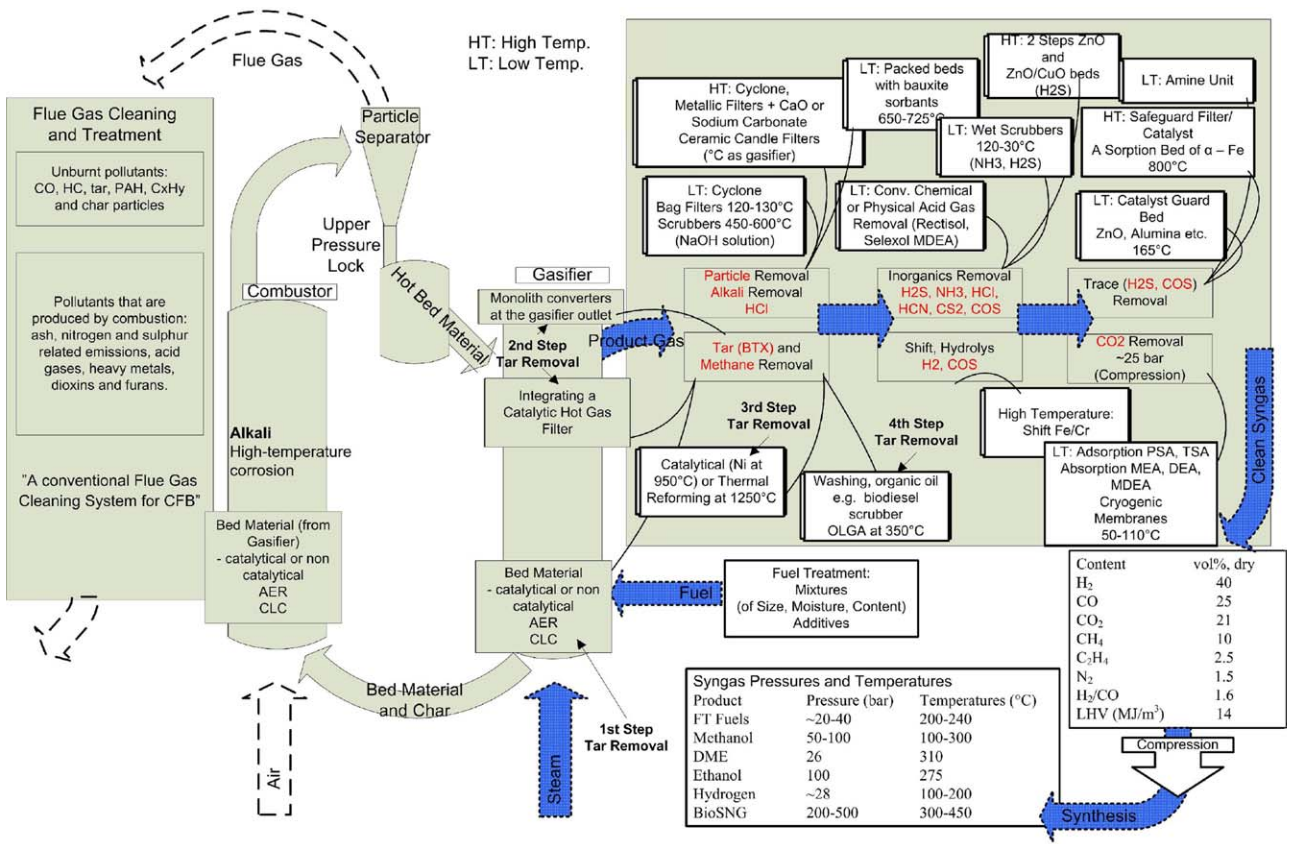


Fig. 2. Overview syngas cleaning and treatment [3,35,40,43,45–50].

A conventional gas purification arrangement for a biomass plant includes:

(1) *Particle, alkali and HCl removal* by cyclones, filters, scrubbers and packed beds with sorbents. Since alkali metals condense at 550 °C on the particulates, they can be removed with particulates.
(2) *Acid gas and inorganic components removal* (including particulates, ammonia, chlorides, other trace organic and inorganic components and residual tars removal) by scrubbers and conventional acid gas removal processes with chemical or physical solvents (such as Rectisol, Selexol MDEA). Chemical solvents for acid gas removal, such as MDEAs (methyldiethanolamine) form chemical bonds between the acid gas (H_2S, CO_2) and the solvent. Physical solvents for instance Selexol or Rectisol remain chemically non-reactive with the gas, and salts formation is avoided. The Selexol process has been used commercially for a long time, and is a dominant acid-gas removal system in gasification projects [34]. Chemical solvents are favourable at low acid-gas partial pressures, whereas physical solvents are favoured at high acid-gas partial pressures [34].
(3) *H_2S and COS removal* by an amine unit and, if necessary, a catalyst guard bed.
(4) *CO_2 removal* by adsorption, absorption or cryogenic membranes [3,34,52].

5.2. Dry hot-gas cleaning

Dry hot-gas cleaning has a potential to be very efficient, clean and reliable. Concerning atmospheric gasification, hot gas cleaning does not improve efficiency, as the subsequent compression requires syngas cooling anyway [53]. Some high temperature cleaning technologies are described below:

(1) *Solid contaminants removal* by ceramic candle filters or metallic filters.
(2) *Fluid contaminants removal* by sorbents [51].
(3) *Trace removal* by a safeguard filter/catalyst, or a sorption bed.

There is no commercial process available for removal of nitrogen compounds from hot gas. They are normally removed by acid gas cleaning at cold temperature. Chlorides and other trace components can also be removed with the dry fly ash [34].

A hot-gas cleaning system was applied by Schweiger and Hohenwarter to clean biomass product gas for fuel cells. First, particulates are removed in a hot gas cyclone and a sinter metal candle; then chlorine is removed in sodium promoted bed, before H_2S by two-steps in ZnO and ZnO/CuO beds. Finally tars are reformed in a two step tar reformer of dolomite and NiO beds [54].

However, Leibold et al. suggest that the particulate filter can be the key component of the optimised concept of high temperature gas cleaning. A particulate filter could in such a case be combined with a catalyst sensitive to sulphur components, i.e. the sorption of the acid gases combined with particulate filtration in an entrained flow process. The sorbent should be dispersed as a fine powder in front of the filter unit and afterwards deposited on the filter elements with the fly ash, and with a periodically re-cleaning of the filter elements. Moreover, H. Leibold et al. suggest a conversion of tar and NH_3 in the syngas by a catalysts layer applied on the clean-gas side of the filter elements, and finally a safeguards filter [40].

5.3. Flue gas

Flue gas pollutants from the CFB combustor of DFBGs can be classified into [50]:

- Unburnt pollutants: CO, HC, tar, PAH, C_xH_y and char particles;
- Pollutants that are produced by combustion: ash (agglomeration, particulate matter), nitrogen (NO_x) and sulphur related emissions (SO_x), acid gases (HCl) and heavy metals. Besides, a certain extent of dioxins and furans.

The first category can be handled with operating variables and combustor design consideration. The second category is significantly related to the properties of biomass and may need secondary measures [50]. A conventional flue gas cleaning system for CFB is commonly used.

6. Tar treatment

Biomass and waste gasification results in higher tar content of the gas compared to coal gasification. The biggest challenge for fluidised-bed gasification is reforming of tars and CH_4 to the minimum allowable limit dependent on the end user application [3].

Various research groups are defining tar differently. According to the ECN definition, tar comprises all organic components having a higher molecular weight than benzene which is not considered to be a tar [55]. Since benzene is one of the major and more stable aromatic compounds and may cause technical or environmental problems, some researchers consider benzene as a tar. In other way, the tars can either be classified in a system focusing on their physical properties (water solubility or condensation) [55,56], or in a system focusing on the process conditions in which the compounds are formed [57] [58].

Tar concentration in biomass syngas is in the order of 10 g/m_n^3 for fluidised-bed gasifiers [58]. However, the fouling problems are not significant as long as all the tar is present in gas phase. Hence, the tar problem is primarily not concerned with the tar quantity, but the properties and the composition of the tar. In condensation-related issues the tar dew point is one of the important parameters. The water solubility of the tar is another important parameter, since the pollution of wastewater is related to the type of tar components being present in the syngas [55].

The reforming of tars and CH_4 to an acceptable low level is achieved by destruction at high temperature, over 1250 °C, or catalytic destruction at low temperature under 950 °C, as well as physical tar treatment like biodiesel scrubbing or the *OLGA* process [3]. These can be divided into two kinds of methods: primary methods and secondary methods. The primary method should be considered before the secondary method. The primary methods are more cost effective, since thermodynamic efficiency losses can be kept to a minimum. An ideal primary method concept would eliminate the use of a secondary method.

6.1. Primary methods

The primary method for controlling the tar content in the syngas means tar prevention and treatments inside the gasifier, such as: the optimising of the gasifier design, the operation of the gasifier, the addition of catalytic bed materials, and the biomass fuel properties. To prevent tar formation, the gasification temperature has a noticeable effect on tar quantity and composition. Also increasing of the gas residence time in a hot zone has a similar effect [58].

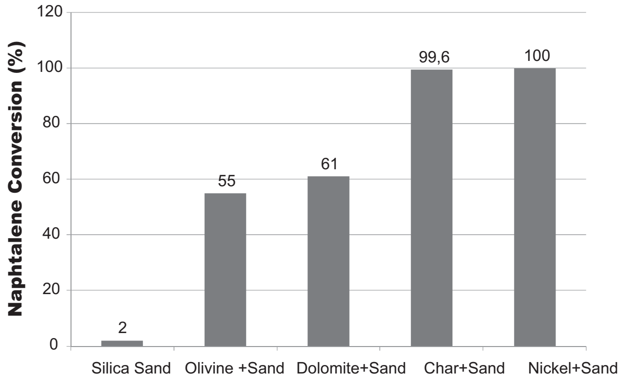


Fig. 3. Effect of catalytic bed material at 900 °C.

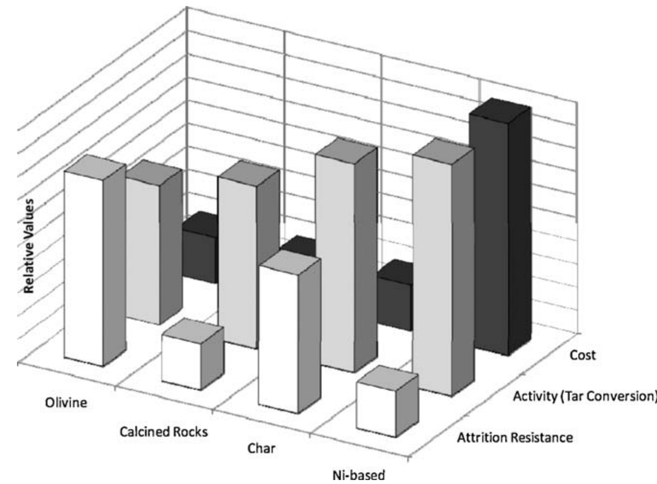


Fig. 4. Catalyst evaluation for biomass gasification (attrition resistance for Ni-based catalyst: deactivation resistance).

6.1.1. Catalytic bed materials

The *first step in tar removal* (see Fig. 2) mainly consists of catalytic bed materials. Examples of natural minerals catalysts are dolomite, limestone, olivine sand, bauxite, natural alumina, clay minerals and iron ore. Some examples of metallic and metal oxide synthetic catalysts are nickel (typically nickel aluminates with lanthanum and cobalt as promoters or nickel enriched natural olivine) and ruthenium based catalyst [48].

The total tar reduction factor at 900 °C is close to the naphthalene reduction, as naphthalene is the dominating compound in the tar at this temperature. The naphthalene conversion at 900 °C is presented in Fig. 3 [59].

6.1.2. Internal reforming of tars

The *second step in tar removal* (see Fig. 2) includes internal reforming of tars by inclusion of a catalytic hot gas filter in the freeboard of the gasifier and at the gasifier outlet. The great reforming activity of a hot gas filter in the gasifier environment has been described in Section 3.2.

6.2. Secondary methods

Secondary methods include cleaning downstream the gasifier, see Fig. 2. Although primary measures are of importance, the combination of proper primary measures with downstream methods is necessary. There are mainly three tar removal systems: thermal cracking, catalytic cracking and mechanical separation.

6.2.1. Thermal and catalytic reforming of tars

The *third step in tar removal* (see Fig. 2) is reforming (thermal and catalytic) of tars. Thermal cracking of tars requires temperatures around 1000–1300 °C. Thermal treatment is cheap and easy to control but leads to heat losses and low efficiency [52]. Catalytic cracking efficiency is 90–95%, and the syngas is simultaneously upgraded. The process needs reaction temperatures of ~800–900 °C [52]. The catalysts can be added to the bed material in a fluidised-bed gasifier, as mentioned above, or in downstream catalytic beds, monoliths or filters.

- *Downstream catalytic beds*: The disadvantages are the cost of the catalysts and the complexity of controlling the process. Natural minerals, char, metallic and metal oxide synthetic catalysts are common materials studied for catalytic bed use [48].
- *Monolith reactors*. Monoliths are ceramic blocks containing a honeycomb structure with a thin layer of catalytically active material on the channel walls, such as a Ni-based coating [48].
- *Catalytic filters*. The catalytic filter combines tar cracking and solids filtration into a single system step [48].

For a summary of the behaviour of catalysts for biomass gasification [60], see Fig. 4. The rapid deactivation on the Ni-based catalyst can be avoided using a DFBG with continuously internal regeneration of the catalyst. It should be noted, the figure is not perfectly according to scale. The purpose is to provide an overview of the attrition resistance, the activity on tar conversion and the cost for different catalysts, compared to each other.

6.2.2. Mechanical separation

The *fourth step in tar removal* (see Fig. 2) is mechanical separation technologies, often applied in combination with catalytic tar removal technologies.

- *Scrubber*. A Scrubber is easy to run and has beneficial effect on air pollution control, but leads to heat losses and wastewater production [52]. The scrubber agent can be water or a scrubbing liquid compatible with tar. The technology of tar scrubbing with oil developed by ECN was given the name OLGA [58].
- *Filters*. The tars are captured on the filter surface, but exist in a sticky liquid form and are hence more difficult to remove from the filter surface than e.g. dust [48].
- *Electrostatic precipitators* (ESPs). A wet ESP is required for biomass gasifier gas tar removal. Tar condensation on dry ESPs would gradually inhibit the particle removal [48].
- *Cyclone separators.* In a cyclone the centrifugal force is used to separate solids and aerosols from the gas. But, the combination of particulates and sticky tar creates a difficult deposition on the cyclone surfaces. Furthermore, cyclones are ineffective at removing small-diameter tar aerosols [48].

7. Conclusions

At present, indirect DFBGs operate near atmospheric pressure. A pressurised DFBG system is worth to consider, for increased gasifier capacity.

A high-quality syngas for automotive fuels synthesis has a strict specification of impurities, since the synthesis catalysts are sensitive to contaminants. The target levels which had to be met for particulate and contaminants are extremely low. The bed material in DFBGs renders in situ gas conditioning. But, although the composition of the syngas is optimised by reactive bed material and the gasification parameters, further treatment is required.

The biggest challenge for fluidised-bed gasification is reforming of tars and CH_4. The primary method for tar treatment should be

considered before the secondary method, because of its inherent potential to be more cost effective.

Dry hot-gas cleaning has a potential to be efficient, clean and reliable—especially since the particulate filter combined with a catalyst can be a key component.

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